

Temperature Dependence of the Magnetic Penetration Depth in the Vortex State of the Pyrochlore Superconductor, $\text{Cd}_2\text{Re}_2\text{O}_7$

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We report transverse field and zero field muon spin rotation studies of the superconducting rhenium oxide pyrochlore, $\text{Cd}_2\text{Re}_2\text{O}_7$. Transverse field measurements ($H=0.007$ T) show line broadening below T_c , which is characteristic of a vortex state, demonstrating conclusively the type-II nature of this superconductor. The penetration depth is seen to level off below about 400 mK ($T/T_c \sim 0.4$), with a rather large value of $\lambda(T=0) \sim 7500$ Å. The temperature independent behavior below ~ 400 mK is consistent with a nodeless superconducting energy gap. Zero-field measurements indicate no static magnetic fields developing below the transition temperature.

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The pyrochlore transition metal oxides, of general formula $\text{A}_2\text{B}_2\text{O}_7$, have been the topic of much interest in recent years as they represent ideal systems for studying the effects of geometrical frustration [1]. Both the A and B sublattices form a network of corner-sharing tetrahedra such that it may not be possible to energetically satisfy all the magnetic interactions simultaneously. The resultant geometric frustration leads to the formation of exotic ground states. Much of the recent work has concentrated on local moment systems where novel properties such as cooperative paramagnetism [2], partial, non-collinear antiferromagnetic ordering [3, 4], spin-freezing [5], and dipolar “spin-ice” behavior [6, 7] have been observed. There has, however, been growing interest in the interplay between itinerant and local moments in geometrically frustrated systems. The metallic pyrochlore $\text{Nd}_2\text{Mo}_2\text{O}_7$ exhibits a large anomalous Hall effect which has been attributed to the Berry phase produced by spin chirality on the pyrochlore lattice [8], while the spinel compound, LiV_2O_4 has been claimed to represent the first known transition metal heavy-fermion system and evidence exists that the unusual properties of this material are related to geometrical frustration on the spinel lattice. [9]

A vast body of work has been carried out on $3d$ and $4d$ transition metal pyrochlores. These are generally insulators and possess either a spin-glass-like or long-range ordered magnetic structure. In contrast, the $5d$ transition metal pyrochlores are mainly metallic, resulting from the extended nature of the $5d$ orbitals. The exception to this is $\text{Cd}_2\text{Os}_2\text{O}_7$ [10] where the $5d^3$ configuration of Os^{5+}

results in a half-filled t_{2g} band and a metal-insulator transition at 226 K. Despite the large number of transition metal compounds which crystallize in the pyrochlore structure and the wide range of physical phenomena observed in these materials, superconductivity had not been observed until the recent discovery of bulk superconductivity in the $5d$ pyrochlore, $\text{Cd}_2\text{Re}_2\text{O}_7$ [11, 12].

$\text{Cd}_2\text{Re}_2\text{O}_7$ crystallizes in the pyrochlore structure with room temperature lattice constant $a=10.219$ Å and an oxygen positional parameter $x=0.3089$. [13] Recent investigations [11, 12, 14, 15, 16] have demonstrated the existence of two phase transitions in this compound. The first, occurring at a temperature of about 200 K, is a continuous structural transition which is accompanied by drastic changes in resistivity and magnetic susceptibility [15, 16]. On further lowering the temperature, $\text{Cd}_2\text{Re}_2\text{O}_7$ has been shown to exhibit bulk superconductivity below a sample dependent transition temperature of about 1 K [11, 12, 14]. Preliminary measurements in the superconducting state indicate that $\text{Cd}_2\text{Re}_2\text{O}_7$ is a type-II superconductor with H_{c1} less than 0.002 T and estimates of the upper critical field, H_{c2} , ranging from 0.2 T to 1 T [11, 12, 14]. None of the measurements reported to date extend below 0.3 K ($T/T_c \sim 0.3$) and hence, little can be concluded about the symmetry of the order parameter in this system. An exponential form of the specific heat as T approaches zero was speculated by Hanawa *et al.* [12] but they point out that measurements to lower temperatures are clearly needed. We report the first measurements on $\text{Cd}_2\text{Re}_2\text{O}_7$ below 300 mK, temperatures which are necessary (for $T_c \sim 1$ K) to extract

information about the superconducting order parameter symmetry. We have performed transverse field (TF) and zero field (ZF) muon spin rotation (μ SR) measurements on single crystal samples of $\text{Cd}_2\text{Re}_2\text{O}_7$. The ZF- μ SR measurements reveal very small internal magnetic fields which are characteristic of nuclear dipoles, indicating no significant electronic magnetism either above or below T_c . The TF- μ SR results provide the first measurement of the internal field distribution in the vortex state in this material. In particular, temperature dependent studies from 20 mK to 4 K indicate a penetration depth which levels off as $T \rightarrow 0$, suggestive of a fully gapped Fermi surface with a rather large zero temperature value of the penetration depth, $\lambda(0) \sim 7500$ Å.

Muon spin rotation has proven to be a very effective probe in the study of superconductivity [17]. In particular, TF- μ SR provides a measure of the length scales associated with type-II superconductors, the penetration depth, λ and the vortex core radius r_0 [17]. In a TF- μ SR experiment, spin polarized muons, with polarization perpendicular to the applied magnetic field direction, are implanted in a sample at a location which is random on the length scale of the vortex lattice. The muon precesses at a rate proportional to the local magnetic field providing a measure of the local field distribution, $n(B)$. The presence of the vortex lattice results in a spatially inhomogeneous field distribution and a resulting muon spin depolarization.

Early TF- μ SR measurements assumed a Gaussian distribution of magnetic fields and with this approximation, the penetration depth can be directly obtained from the Gaussian depolarization rate, $\sigma \sim 1/\lambda^2$. This approximation has been shown to be reasonable for the case of polycrystalline samples but is inadequate for the case of single crystals [17]. In this case, a Ginzburg-Landau (GL) model has been developed to model the magnetic field distribution for a single crystal. In GL theory, the size of the vortex core is determined by the applied magnetic field, H , and the GL coherence length normal to the applied field, ξ_{GL} , while the penetration depth provides the length scale of the decay of magnetic field away from the vortex core. The field distribution is calculated from the spatial distribution of magnetic field [18],

$$B(\mathbf{r}) = \frac{\Phi_0}{S}(1 - b^4) \sum_{\mathbf{G}} e^{-i\mathbf{G} \cdot \mathbf{r}} \frac{u K_1(u)}{1 + \lambda^2 G^2}, \quad (1)$$

where $u^2 = 2\xi_{GL}^2 G^2(1 + b^4)[1 - 2b(1 - b^2)]$, $K_1(u)$ is a modified Bessel function, \mathbf{G} is a reciprocal lattice vector of the vortex lattice, $b = H/H_{c2}$ is the reduced field, Φ_0 is the flux quantum and S is the area of the reduced unit cell for a hexagonal vortex lattice.

Single crystals of $\text{Cd}_2\text{Re}_2\text{O}_7$ were grown using vapor-transport techniques as described elsewhere [14, 19]. Three samples with an approximate surface area of 5×5 mm² each were mounted using low temperature grease

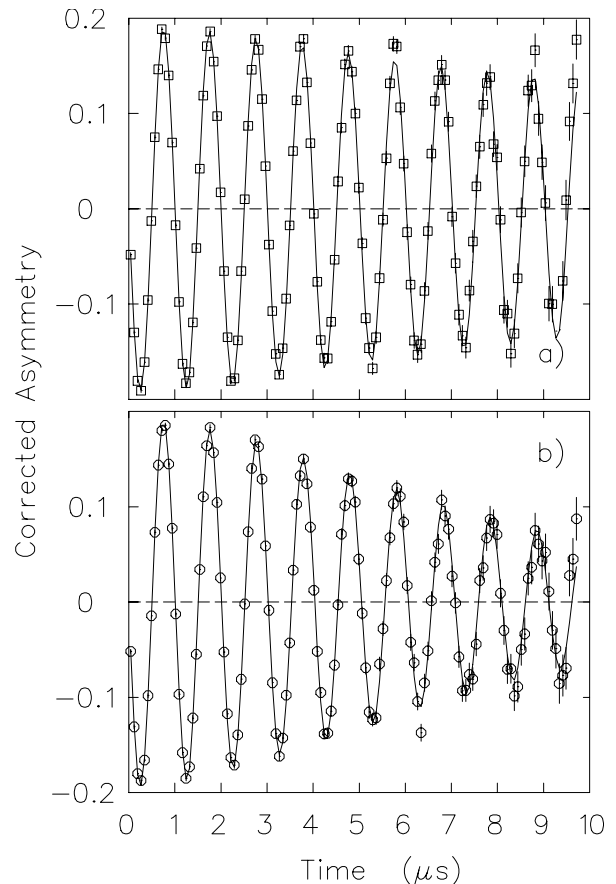


FIG. 1: Typical μ SR spectra in $\text{Cd}_2\text{Re}_2\text{O}_7$ obtained in a transverse magnetic field of 0.007 T at temperatures of (a) $T=1.5$ K (above T_c) and (b) $T=100$ mK (below T_c).

such that the cubic (100) direction would be parallel to the applied magnetic field direction. They were mounted on intrinsic GaAs in order to eliminate any precession signal at the background frequency [20] from muons which miss the sample and would otherwise land in the Ag sample holder. The samples were covered with 0.025 mm Ag foil which was bolted to the sample holder to ensure temperature uniformity. The TF and ZF- μ SR measurements were performed in an Oxford Instruments dilution refrigerator on the M15 beamline at TRIUMF at temperatures from 20 mK up to 4 K.

Given the estimated critical field values, we selected a field value of 0.007 T and the temperature dependence was measured by cooling the sample in the presence of this applied magnetic field to ensure a uniform flux line lattice. Figs. 1(a) and (b) show typical μ SR spectra in a transverse field of 0.007 T, for temperatures above and below T_c respectively. Examination of this data clearly shows an enhanced depolarization rate on entering the superconducting state resulting from the inhomogeneous field distribution associated with the flux line lattice. This represents the first experimental observation of the vortex lattice in $\text{Cd}_2\text{Re}_2\text{O}_7$ and provides clear evidence

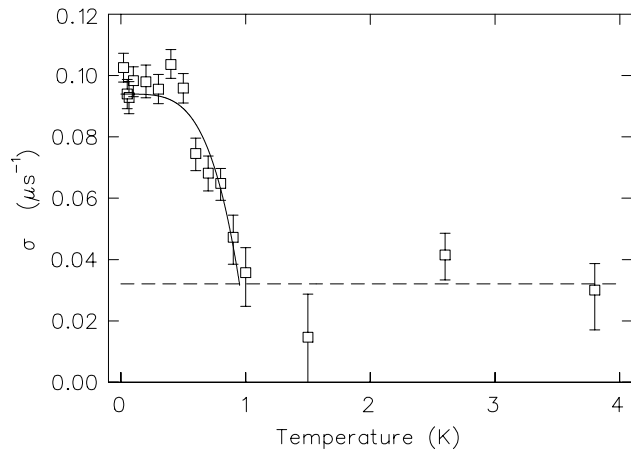


FIG. 2: Linewidth parameter σ as a function of temperature in a transverse magnetic field of 0.007 T applied along the (100) direction. The solid line is a fit to Eq. 2.

that this material is a type-II superconductor. The observed increase in the TF line broadening below T_c can be attributed entirely to the vortex lattice since the ZF muon spin relaxation rate (not shown here) was small and roughly temperature independent below 2 K.

The solid lines shown in Figs. 1(a) and (b) represent fits of the individual time spectra to a sample signal consisting of a Gaussian envelope with fixed asymmetry and a background signal with fixed linewidth and asymmetry. The background, from muons which miss the sample and land in the heat shields, was obtained independently by performing measurements with the sample removed. The resulting sample linewidth, σ , is shown in Fig. 2 as a function of temperature. As one can clearly see, the magnitude of the depolarization rate as $T \rightarrow 0$ is very small, saturating at a value of about $0.1 \mu s^{-1}$. The residual linewidth from nuclear dipoles, taken from the data above T_c , is very small in $Cd_2Re_2O_7$ (about $0.03 \mu s^{-1}$), allowing for clear observation of the line broadening associated with the flux line lattice. Apparently the muons stop in sites which are not close to the Re ions, which have appreciable nuclear moments.

As can be seen from Eq. 1, the field distribution depends on both the penetration depth and GL coherence depth. The GL coherence length can be obtained from the known value of the upper critical field using the expression $\xi_{GL} = (\Phi_0/2\pi H_{c2})^{1/2}$ where Φ_0 is the flux quantum. As mentioned above, a range of values for H_{c2} have been reported and consequently, to provide a self-consistent measurement of λ , the field dependence of the linewidth was measured. To account for any possible instrumental field-dependence in the linewidth, measurements were made above the transition temperature (2 K) at each field value after which the sample was field-cooled to 100 mK. The measured linewidth in the normal state was subtracted in quadrature from that ob-

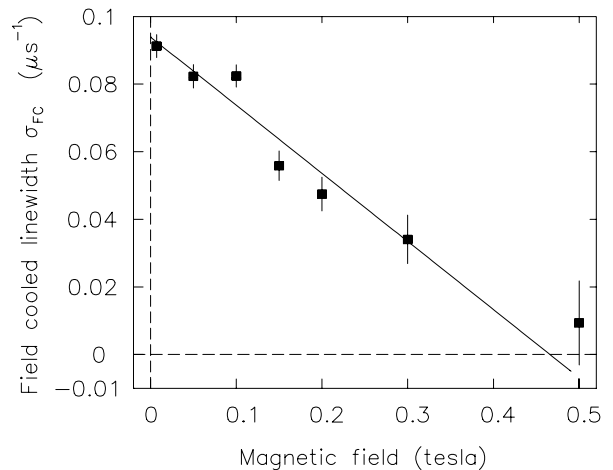


FIG. 3: Linewidth parameter σ_{FC} as a function of magnetic field applied along the (100) direction. The normal state contribution has been subtracted as described in the text. Measurements were taken at a temperature of 100 mK.

served at 100 mK and the results are plotted in Fig. 3 as a function of applied magnetic field. As can be clearly seen, the linewidth decreases almost linearly with applied field. This is attributed to the linear increase in the volume taken up by the vortices. The linewidth parameter, σ_{FC} , approaches zero at a field of 0.5 T which is our estimate of $H_{c2}(T \rightarrow 0)$ and is consistent with measurements on other samples. This estimate of the critical field corresponds to $\xi_{GL} \sim 260 \text{ \AA}$. Using this value, we obtain the penetration depth using the field distribution shown in Eq. 1. The resulting temperature dependent penetration depth is shown in Fig. 4. As expected from the small values of linewidth, at the base temperature we observe a rather large value of the penetration depth, $\lambda(0) \sim 7500 \text{ \AA}$. We note that this value of penetration depth is significantly larger than most oxide superconductors where values ranging from 1000-2000 \AA are typical [21, 22].

As the penetration depth is related to the concentration of superconducting carriers, its temperature dependence is a measure of the low-lying electronic excitations. As such, the presence of a nodeless superconducting energy gap is indicated by a leveling off of the penetration depth as the temperature decreases below T_c . As is clearly seen in Figs. 2 and 4, the linewidth and penetration depth respectively become temperature independent as the temperature decreases below about 0.4 K, consistent with a fully gapped Fermi surface. Consequently, we conclude that the superconducting order parameter in $Cd_2Re_2O_7$ is consistent with a nodeless energy gap suggesting either s -wave symmetry or exotic pairing symmetries, such as p -wave which can also exhibit a fully gapped Fermi surface. For comparison, the solid line in Figs. 2

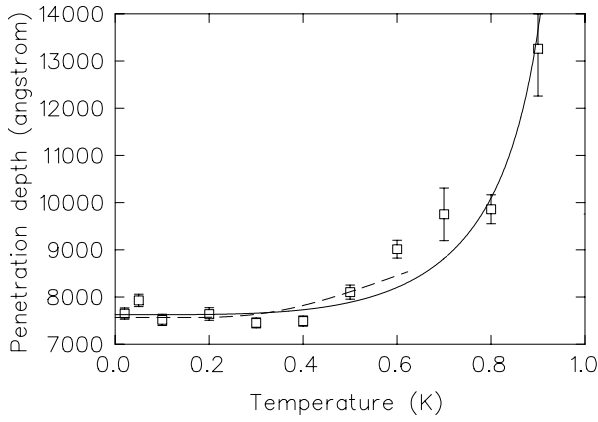


FIG. 4: Penetration depth as a function of temperature in a magnetic field of 0.007 T applied parallel to the (100) direction. The solid line is a fit to Eq. 2.

and 4 represent fits to the two fluid approximation

$$\frac{\sigma(T)}{\sigma(0)} \sim \frac{\lambda^2(0)}{\lambda^2(T)} = [1 - (T/T_c)^4], \quad (2)$$

while the dashed line in Fig. 4 is a fit to the BCS temperature dependence

$$\lambda(T) = \lambda(0) \left[1 + \sqrt{\frac{\pi\Delta_0}{2T}} \exp\left(\frac{-\Delta_0}{T}\right) \right] \quad (3)$$

where $\Delta_0 = 1.74(11)$ K.

The London penetration depth, λ , provides a direct measure of the ratio of superconducting carrier concentration to effective mass, n_s/m^* ,

$$\frac{1}{\lambda^2} = \frac{4\pi n_s e^2}{m^* c^2} \left(1 + \frac{\xi_0}{l} \right)^{-1}, \quad (4)$$

where ξ_0 is the Pippard coherence length, and l is the mean-free path. There is considerable uncertainty in estimations of the mean-free path with reported values ranging from 200-700 Å [14, 23] and it is unclear whether $\text{Cd}_2\text{Re}_2\text{O}_7$ is a superconductor in the clean or dirty limit. If the material is in the clean limit, the present results provide strong evidence for a fully gapped Fermi surface. Under the assumption of a clean superconductor, such that $\xi_0/l \ll 1$, a value of $n_s m_e/m^*$ of $5.0 \times 10^{25} \text{ m}^{-3}$ can be obtained using Eq. 4 and the measured penetration depth. On the other hand, if $l \sim 200$ Å (i.e. the dirty limit) then we obtain $\xi_0 \sim 470$ Å and $n_s m_e/m^* \sim 1.4 \times 10^{26} \text{ m}^{-3}$. Clearly, precise determination of the mean free path for $\text{Cd}_2\text{Re}_2\text{O}_7$ is needed to allow accurate quantitative information to be extracted.

In conclusion, we have performed μSR studies of the superconducting state in the recently discovered pyrochlore superconductor, $\text{Cd}_2\text{Re}_2\text{O}_7$. Zero-field measurements indicate no significant magnetism in this superconductor, suggesting that magnetic frustration does not

play a direct role in the superconductivity. Transverse-field measurements show that $\text{Cd}_2\text{Re}_2\text{O}_7$ is a type-II superconductor and indicate a superconducting order parameter consistent with a fully gapped Fermi surface with a zero temperature value of penetration depth of ~ 7500 Å. However, considering that the superconductor may be in the dirty limit, spectroscopic techniques which directly measure the density of states would be required to confirm this conclusion.

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